

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of:

Confirmation No.: 7418

Frank Henglein, *et al.*

Serial No.: 10/580,120

Group Art Unit: 1793

Filed: September 11, 2006

Examiner: Pegah Parvini

For: EFFECT PIGMENTS HAVING AN ALUMINUM OR ALUMINUM ALLOY  
CORE, PROCESS FOR THE PRODUCTION THEREOF AND USE THEREOF

VIA EFS-WEB

Commissioner for Patents

P.O. Box 1450

Alexandria, VA 22313-1450

SECOND §1.132 DECLARATION OF DR. FRANK HENGLEIN

Sir:

I, Frank Henglein, Ph.D., do hereby declare as follows:

1. I am the same Dr. Frank Henglein who previously authored a Declaration under 37 C.F.R. §1.132 dated September 16, 2008 which, I am informed, was filed with the U.S. Patent Office on September 17, 2008 as an Attachment to an Amendment in response to the March 18, 2008 Office Action concerning this application. My qualifications are as set forth in the previous declaration. This supplemental declaration is being provided to respond to issues raised by the Examiner in her subsequently issued final Office Action, dated December 31, 2008, with regard to the comparison described in my previous declaration between pigments produced by a wet chemical oxidation process according to claim 1 of this application and those produced by a wet chemical coating process according to the disclosure contained in U.S. Patent No. 5,624,486 to Schmid et al. ("Schmid").
2. In the present Office Action (dated December 31, 2008) the Examiner continues to maintain her "obviousness" rejection of claims 1-36 and 39-43 under 35 U.S.C. §103 from the March 18, 2008, Office Action based on the combination of the Schmid patent

with U.S. Patent No. 6,648,957 to Andes et al. ("Andes"). As mentioned above, in my first Declaration I compared the structure and characteristics of an aluminum effect pigment produced with the use of a wet chemical oxidation technique as described in claim 1 of this application with those of an effect pigment produced in accordance with the wet chemical coating process described in the 'primary' (i.e., Schmidt) reference relied upon by the Examiner to reject the claims. The purpose of the comparison was to demonstrate the unique structure and features of a pigment produced by the claimed methodology in accordance with, e.g., the recitation of claim 1.

3. As set forth in my previous declaration, the outer surface of the pigment recited in claim 1 of the present application is roughened in that its structure comprises dendritic elevations and depressions. This structure provides the claimed pigment with a soft color flop (see ¶5 of my prior declaration). This is in contrast with a pigment produced according to Schmidt et al., as also noted in my prior declaration, which has a granular surface structure without the surface roughening found in the claimed pigment(s), which surface produces a strong color flop (see ¶8 in my previous declaration). As summarized in ¶10 of my prior declaration, therefore, the pigments according to the present claims have an entirely different structure than those produced according to the teachings contained in Schmid. Due to this difference in structure, moreover, these pigments differ substantially from one another with regard to their color flop properties, with our claimed pigment having the more desirable "soft" color flop property in comparison to a pigment particle produced according to Schmidt.

4. In the December 31, 2008 Office Action, however, the Examiner raised several objections to my first declaration (see, e.g., p. 3 of the Office Action). From the remarks provided by the Examiner I understand that she is alleging that my previous declaration does not set forth a proper comparison between the pigment as presently claimed in this case versus a pigment produced with a wet chemical coating process in accordance with the teachings contained in the Schmidt et al. U.S. patent No. 5,624,486, and further, that she is alleging the declaration is not commensurate with the scope of the claims, i.e., in

that claim 1 does not recite the color flop feature noted above. The following remarks are, therefore, provided to address the Examiner's objections.

5. I understand that my Counsel has amended claim 1 of the present application in an Amendment to be filed in company with this declaration in order to further clarify the distinguishing characteristics of the claimed effect pigments. In particular, I understand that claim 1 is amended in the accompanying Amendment such that it now includes, first, a recitation that the pigments have a weak color flop having a  $\Delta H^*_{\text{anchor}}$  in a range of between 1.5 to 50, as taught for example at p. 10, lines 1-2 of the present application. Furthermore, I additionally understand that the subject claim has also been further amended to recite, as originally set forth in claim 13, i.e., that the at least one highly refractive metal chalcogenide layer having a refractive index of  $> 1.95$  comprises a colored metal chalcogenide layer or a plurality of colored metal chalcogenide layers. The impact of the features which have been added to claim 1, in terms of how they serve to further distinguish the presently claimed effect pigments from those described in the prior art, e.g., in the Schmid reference, is discussed below.

6. Regarding the weak color flop feature now incorporated into claim 1 the Examiner's attention is respectfully directed to the paragraph set forth at the top of p. 10 in the present application. In the indicated paragraph the color location value  $H^*$  is explained as the angle of the color location vector to the positive  $a^*$  axis. The color flop  $\Delta H^*_{\text{anchor}}$  is the quantitative maximum difference of the individual  $H^*_{\text{anchor}}$  values of the five anchor angles  $25^\circ/140^\circ$ ;  $45^\circ/150^\circ$ ;  $45^\circ/120^\circ$ ;  $75^\circ/120^\circ$  and  $75^\circ/90^\circ$  (see the application at p. 26, lines 19 - 21).  $\Delta H^*_{\text{anchor}}$  values for pigments with no substantial or no color flop and  $\Delta H^*_{\text{anchor}}$  values for pigments with a steep color flop are also set forth in the indicated paragraph. Furthermore table 4 at p. 27 of the application shows a comparison of  $\Delta H^*_{\text{anchor}}$  values between the pigments claimed in the application and the commercial market products Variocrom (Comp. Ex. 9) and Paliochrom (Comp. Ex. 11) which are both available from BASF. The steep color flop of Variocrom with a  $\Delta H^*_{\text{anchor}}$  value of 77.05 differs significantly from a pigment with no color flop such as Paliochrom with a  $\Delta H^*_{\text{anchor}}$  value of 1.00. The  $\Delta H^*_{\text{anchor}}$  values of the pigments according to claim 1

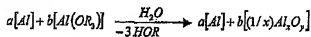
(Ex. 1, 2 and 4) are in between these values, therefore they are in the range for pigments exhibiting a weak color flop. Comp. Ex. 7 (Aloxal) is an oxidized aluminum pigment according to example 1 without subsequent coating with iron oxide. For a better representation of the color flop, under the designation "effect anchor" the  $a^*, b^*$  color values were plotted against each other (see Fig. 7 and for a description of Fig. 7, see p. 26 of the application). The effect anchor represents a measure of the color flop. The higher the  $\Delta E^*_{\text{anchor}}$ , the greater the shift in the color space and the color flop resulting thereof. In Fig. 7 Varlocrom with the steepest flop in this comparison exhibits the greatest changes in the color location at different observation angles. The effect anchor is strongly pronounced. In contrary Pallocrom with no color flop forms no effect anchor, the effect anchor is a prolongation of the effect line. The pigments according to the present invention (Ex. 1, 2, 4), i.e., having a weak color flop, are different from those described above. The effect anchor spreads only to a moderate extent in the color space. Further to the above, moreover, all the examples of Schmid et al. are qualitatively described with respect to their color flops. From this qualitative description one such as myself who is skilled in this art can readily deduce  $\Delta E^*_{\text{anchor}}$  values much higher than 50 units for all of the examples set forth in the subject reference.

7. Turning next to a substantive discussion of the differences, i.e., in structure and characteristics, between the effect pigments produced via the wet chemical oxidation process in accordance with claim 1 of the present application and those (pigments) produced with the use of a wet chemical coating process according to, e.g., U.S. Patent No. 5,624,286 to Schmid et al., I submit that the two processes are completely different. The wet chemical coating is a kind of an exogenously generated coating by hydrolytic decomposition of organic (...) compounds (Schmid et al., col. 13, lines 6 - 10). This leads to distinct layers, which are applied one after another. The wet chemical oxidation process as used in forming our claimed pigments, on the contrary leads to a partial conversion of the aluminum substrate to aluminum oxide, i.e. the aluminum oxide on the aluminum substrate is produced by a directed oxidation of the aluminum substrate. Thus the thickness of the substrate metal pigment is reduced significantly after the oxidation

process whereas it is not changed during the wet chemical deposition of e.g.  $Al(OH)_3$  described by Schmid et al.

8. The following reaction equations quantitatively emphasize the above mentioned fundamental difference between a "wet chemical coating" and a "wet chemical oxidation" process (in accordance with the equation set forth on p. 8, line 22 of the present application):

Wet chemical coating:



Wet chemical oxidation:



Regarding the disclosure of the percentage by weight of the aluminium in the core of the resulting pigment, I have compared the ratios of aluminium and aluminium oxide. Percentage by weight and ratio are directly related to each other. In the case of the wet chemical coating (i.e., according to Schmid et al.) a ratio of  $a/b$  is seen, whereas the ratio is  $(a-b)/b$  in the case of the wet chemical oxidation technique recited in claim 1. According to claim 1 the ratio  $(a-b)/b$  is less than 90 % by weight. Therefore the amount of aluminium in likewise obtained pigments in case of the wet chemical oxidation is clearly smaller compared to the wet chemical coating process. This comparison thus clearly demonstrates that there is a substantial difference between the wet chemical oxidation process recited for use in claim 1 and the wet chemical coating process disclosed in the Schmid et al. reference.

9. Further with regard to the reaction equations set forth in ¶7 above, the ratio of the aluminium oxide in the case of the wet chemical coating is defined by  $b/(a+b)$ . In the case of a wet chemical oxidation, however, it is  $b/a$ . Therefore the amount of aluminium oxide

related to that of aluminium in a pigment produced by the wet chemical oxidation is generally higher and the thickness of the central layer of a metal pigment is lower compared to a pigment produced by wet chemical coating.

10. A further important difference between the wet chemical coating process of Schmid et al. and the wet chemical oxidation process recited in, e.g., claim 1 of the present application lies in the fact that according to the teaching of Schmid, organic metal compounds are added to the reaction solution (see, e.g. col. 5, line 62 to col. 6, line 42). The organic metal compounds are immediately hydrolysed upon contact with water. As a result metal oxide/hydroxide, such as aluminium oxide/hydroxide, is formed and precipitated on the metal pigments. The deposition of the precipitating metal oxide/hydroxide on the metallic substrate produces a protecting layer and, consequently, prevents an oxidation of the metallic substrate (e.g. aluminium pigment). Therefore, contrary to the pigments according to, e.g., claim 1, the surface of an aluminium pigment is not roughened (oxidized) when following the teaching of Schmid. In the absence of the organic compound  $Al(OR)_3$ , water will react with the aluminium flakes, if a basic or acidic catalyst is present leading to the wet chemical oxidation. In fact, I have observed that the wet chemical oxidation process can be stopped by adding e.g.  $Si(OCH_2CH_3)_4$  to the reaction mixture.

11. With respect to the layer roughness, this issue was already discussed in, e.g., ¶5 of my prior declaration. The enhanced layer roughness is a consequence of the use of the wet chemical oxidation technique recited in claim 1 of the present application. The aluminium oxide produced by the direct oxidation of the substrate exhibits a relatively rough surface which is not obtainable by a wet chemical coating. If a further layer is applied to the aluminium oxide layer obtained by wet chemical oxidation, this layer will be incorporated into the rough and porous surface of the aluminium oxide. In this way a mixed layer of aluminium oxide and the further layer is obtained (cf. patent application as filed, p. 9, line 6 - 9).

12. The aluminium oxide layer is restricted in claim 1 to having a thickness of from 50 to 300 nm. In the case of an interference pigment having a semitransparent metal layer or a black metal oxide layer with a high refractive index on the aluminium oxide layer and distinct layers as described by Schmid, such thicknesses may produce interference colors, depending on the thickness of the aluminium oxide layer. Furthermore the interference pigments exhibit color travel, because the light seen at different angles has to travel a different optical way through the layer stack. A further increase, i.e., to a thickness above 300 nm, would lead to even more pronounced colors as higher order interference colors are produced. In the case of the rough aluminium oxide obtained by the wet oxidation process, however, still different colors can be produced by changing the thickness of the aluminium oxide layer. The dependence of the interference color on the thickness, is however, much less pronounced than in the case of distinct layers. The mixed layer formed by the metal chalcogenide and the pores of the aluminium layer form an intermediate layer of a refractive index being in between the high and the low refractive index layers. An increase of the thickness of the aluminium oxide layer also leads to an increase of this mixed layer. This results in a pigment with essentially no or only a weak color flop, as is now recited in (amended) claim 1 of the present application.

13. In contrast, moreover, to the pigments as recited in claim 1 of the present application, all of the examples of Schmid *et al.* refer to a pigment having a first layer A consisting of aluminum oxide or  $\text{SiO}_2$  and a second layer B consisting of metal or non-selectively absorbing metal oxide (see col. 2, line 67 to col. 3, line 3, or col. 4, lines 26 – 33) and if desired a third layer. The layer B, being metal layer or metal oxide layer, (col. 4, lines 30 – 33) is black (col. 4, line 17 or line 30). The effect of the black layer is explained at col. 4, lines 39-43. By absorbing the white light, the interference colors and consequently the color flop is enhanced, such that the Schmid pigments demonstrate a strong color flop.

14. Still further, by applying a layer of colored metal chalcogenide as is now recited in amended claim 1, light impinging on the effect pigment is selectively absorbed. This is

contrary to the teaching of Schmid et al., according to which the metal oxide must be non-selectively absorbing.

15. In summary, as noted in ¶3 above and in ¶10 of my prior declaration, the pigments produced according to claim 1 of the present application, i.e., with the use of a wet chemical oxidation process, have a completely different structure – manifesting itself in a completely different appearance – than pigments formed according to the wet chemical coating process as disclosed in the Schmidt et al. reference. Due, moreover, to their differences in surface structure, the pigments differ substantially with regard to their properties (e.g., color flop), wherein the pigments according to the present invention have a soft interference color flop, whereas the pigments produced according to Schmidt et al. have a strong color flop.

16. I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: 04/29/09

By:

  
Dr. Frank Henglein